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Primary particle size distribution of eroded material affected by degree of aggregate slaking and seal development

D. N. Warrington[a,b], A. I. Mamedov[c], A. K. Bhardwaj[d] & G. J. Levy[a]

[a]*Department of Soil Chemistry, Plant Nutrition and Microbiology, Institute of Soil, Water and Environmental Sciences, Agricultural Research Organization, The Volcani Center, Bet Dagan 50250, Israel,* [b]*State Key Laboratory of Soil Erosion and Dryland Farming on the Loess Plateau, Institute of Soil and Water Conservation, CAS & MWR, Yangling, Shaanxi Province, 712100, China,* [c]*USDA-ARS-GMPRC-WERU, 1515 College Avenue, Manhattan, KS 66502, USA, and* [d]*Great Lakes Bioenergy Research Center (GLBRC), W.K. Kellogg Biological Station, Michigan State University, 3700 E. Gull Lake Drive, Hickory Corners, MI 49060-9516, USA*

Summary

Primary particle size distribution (PSD) of eroded sediment can be used to estimate potential nutrient losses from soil and pollution hazards to the environment. We studied eroded sediment PSDs from three saturated soils, packed in trays (20 × 40 × 4 cm), that had undergone either minimal aggregate slaking (MAS) or severe aggregate slaking (SAS) prior to a 60 mm simulated rainstorm (kinetic energy, 15.9 kJ m^{-3}; droplet diameter, 2.97 mm) and collected runoff at regular intervals. The degree of aggregate slaking was controlled by the rate at which soils were wetted to saturation. The PSDs of eroded materials and of parent soils were determined using a laser particle size analyser. For each soil, PSD frequency curves of eroded sediments and parent soils were generally of a similar shape but most eroded sediments had larger clay contents than their parent soils. In the SAS treatment, cumulative clay enrichment in the eroded materials was inversely related to the parent soil clay content, these being 28.5, 26.6 and 22.8% richer in clay than their parent soils for the loam, sandy clay and clay, respectively. Generally, total clay loss was greater from soils with SAS than from those with MAS because of erosion rates; however, clay enrichment of sediments, compared with parent soil clay contents, was mostly greater in samples with MAS. Greater clay enrichment took place during the early seal development stage in the loam, but could not readily be associated with specific stages of seal development for the clay. In the sandy clay, the relation between seal development and clay enrichment in the eroded material depended on the initial degree of aggregate slaking. The observed large preferential loss of clay by erosion in cultivated soils re-emphasizes the need to employ erosion control measures.

Introduction

Loss of soil in overland flow is a serious problem worldwide, not only because a non-renewable resource is being lost but also because eroded sediments are a potential source of pollution, degrading water quality in river systems and contaminating downstream areas (Young & Onstad, 1978; Ghadiri & Rose, 1993).

Soil erosion by water involves two main processes: (i) detachment of soil material from the soil mass by raindrop impact and/or runoff shear, and (ii) transport of the resulting sediment by raindrop splash and/or flowing runoff water. Raindrop detachment is greater than flow shear detachment from an exposed soil surface because the kinetic energy of raindrops is much larger than that of surface flow (Hudson, 1971). However, movement of detached soil down-slope by rain splash is minimal, and most of the sediments are removed by overland flow (Young & Wiersma, 1973). Under dispersive conditions (e.g. sodic soils; low electrical conductivity of applied water, either by irrigation or rainfall; soils containing high levels of water dispersible clay minerals) (Norton, 1987), overland flow alone may be sufficient for detachment and transport (Mamedov *et al.*, 2002). Furthermore, detachment by overland flow may increase sharply with slope and when rilling is initiated (Warrington *et al.*, 1989; Shainberg *et al.*, 1992).

Runoff is initiated or enhanced by seal formation at soil surfaces. Seal formation in soils exposed to raindrop impacts results from two mechanisms (Agassi *et al.*, 1981): (i) physical

Correspondence: G. J. Levy. E-mail: vwguy@volcani.agri.gov.il
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disintegration of surface aggregates and their compaction, and (ii) a physico-chemical dispersion and movement of clay and other fine-sized particles down the profile to 0.1–0.5 mm depth, where they may accumulate and clog conducting pores. With the first mechanism, disintegration of soil aggregates at the soil surface is a consequence of both the wetting process (Kemper & Koch, 1966; Loch, 1994; Kay & Angers, 1999) and raindrop impact (Betzalel *et al*., 1995). The degree of aggregate disintegration by raindrops depends on the impact energy of the drops (e.g. Betzalel *et al*., 1995; Mamedov *et al*., 2000), while the degree of aggregate disintegration by wetting depends on the rate of wetting (Quirk & Panabokke, 1962; Loch, 1994). Explosions, caused by entrapped air and differential swelling, were suggested as the main mechanisms for aggregate disintegration by wetting (Panabokke & Quirk, 1957; Quirk & Panabokke, 1962). The extent of aggregate disintegration by wetting, termed 'slaking', also depends on aggregate stability, which is directly related to organic matter, sesquioxides and clay contents (Kemper & Koch, 1966; Kay & Angers, 1999; Norton *et al*., 2006). Shainberg *et al*. (2003) reported that seal formation and runoff amount were enhanced when soils were exposed to fast wetting, and thus to a greater degree of aggregate slaking, before a rain event, compared with when they were exposed to slow wetting, and that this effect increased with aggregate stability.

While there is a clear relationship between seal development and runoff, the relationship between seal formation and soil inter-rill erosion is not so obvious (Assouline & Ben-Hur, 2006). For example, seal formation may affect inter-rill soil erosion in the following counter-acting ways: (i) seal development increases the shear strength of the soil surface (Bradford *et al*., 1987), and thus reduces soil detachment (Moore & Singer, 1990), but (ii) seal formation increases runoff and thus its flow shear force, which in turn increases the transport capacity for entrained material (Moore & Singer, 1990). Similarly, rainfall splash is initially the dominant mechanism for detachment (Hudson, 1971) and as runoff begins, raindrop impact increases the turbulence of the flowing surface water, and hence the erosive power of overland flow (Bradford *et al*., 1987). However, due to further seal formation and development and runoff generation, the depth of water covering the soil surface increases and absorbs the impact of the raindrops (Ferreira & Singer, 1985). Surface seal formation may also be impeded in certain soils because of the presence of deposited soil particles that 'shield' the underlying soil (Hairsine *et al*., 1999). Thus, it is possible to have a situation where the combination of the overland flow and raindrop impact driven erosion processes are either greater or less than the sum of their individual contributions and that this will depend upon the soil properties, the degree of seal development, surface water depth and other factors (Rouhipour *et al*., 2006; Asadi *et al*., 2007).

In order to estimate the effects of the erosion process, it is important to quantify aggregate and primary particle size distributions (PSD) of the eroded sediments in addition to the total amount of soil material lost. Clay is usually considered to be the size fraction of the sediment that is most important in transporting adsorbed chemicals in the soil (Young & Onstad, 1978), although it should be noted that adsorption is non-uniformly distributed among clay particles depending upon their location within a soil aggregate (Ghadiri & Rose, 1993), as well as representing a nutritional loss from the parent soil (Smith *et al*., 2005). Because finer material can travel greater distances before being deposited, an understanding of the range of the PSD of eroded sediments is desirable in order to predict accurately where soil components will be deposited, and how the PSD of the eroded sediment may vary depending on soil type, texture and management practices.

A number of studies have tried to characterize eroded sediments in terms of their effective size distributions (i.e. usually consisting of a mixture of aggregated material and individual soil particles), and/or their primary PSD, determined following complete dispersion of the eroded material, and to compare them with those of the parent soil. The results, however, have varied. Some studies reported that sediments from interrill erosion were enriched in sand at the expense of the silt and clay size fractions (Young & Onstad, 1978; Alberts *et al*., 1980). In other studies it was observed that clay, and not sand, was enriched in the eroded sediment (Monke *et al*., 1977; Alberts *et al*., 1983). It has also been noted that the composition of sediments changes during the rainfall; at the beginning of the storm (i.e. before a surface seal develops) sediments had finer material than that of the parent soil, but at the end of the storm (i.e. when the seal formation process was completed) the PSD of the sediments was comparable to that of the parent soil (Gabriels & Moldenhauer, 1978; Mitchell *et al*., 1983). These differences in reported PSDs of eroded materials relative to their parent soils may arise from differences in their soil properties (e.g. texture, clay content, etc.) and the conditions existing at the soil surface prior to a rainfall event (e.g. the condition of surface aggregates, moisture content and size), as well as the characteristics of the rainfall event itself (e.g. intensity, raindrop size and energy, and duration).

In the current study, it was hypothesised that the primary PSD of the eroded sediments would vary depending on: (i) the degree of slaking of surface aggregates, (ii) the degree of seal development, and (iii) the parent soil clay content. The objective of our study was thus to verify these hypotheses and was an extension of the work of Shainberg *et al*. (2003), who examined only total soil losses.

Materials and methods

We used three of the soils studied by Shainberg *et al*. (2003), a loam (Calcic Haploxeralf), a sandy clay (Chromic Haploxerert) and a clay (Typic Haploxerert), and closely followed their rainfall simulator procedure. Selected properties of the disturbed top layer (0–250 mm) of these typical smectitic soils in Israel are presented in Table 1.

Table 1 Some physical and chemical properties of the soils studied

		Particle size distribution[b] /%									
		Sand		Silt		Clay					
Soil type[a]	Classification	HY	LA	HY	LA	HY	LA	CEC /cmol$_c$ kg^{-1}	ESP /%	$CaCO_3$ /g kg^{-1}	OM /g kg^{-1}
Loam	Calcic Haploxeralf	41.3	40.8	36.2	45.2	22.5	14.0	17.7	2.1	182	12
Sandy clay	Chromic Haploxerert	46.5	10.4	15.4	54.2	38.1	35.4	34.8	1.6	96	11
Clay	Typic Haploxerert	25.0	2.2	13.8	54.2	61.2	43.6	65.0	1.0	50	17

[a]Soil type based on particle size distribution determined by the hydrometer method (Gee and Bauder, 1986); size fractions as defined by the International Soil Science Society.
[b]Particle size analysis determined by the hydrometer (HY) and laser (LA) methods (for comparison).
CEC = cation exchange capacity.
ESP = exchangeable sodium percentage.
OM = organic matter.

Rainfall simulation procedure

Air-dried soil samples, crushed and passed through a 4 mm sieve, were uniformly packed in trays (20 × 40 × 4 cm) and then saturated from the bottom with tap water (electrical conductivity, 0.9 dS m^{-1}; sodium adsorption ratio, 2.5) at either a slow (2 mm hour^{-1}) or a fast rate (64 mm hour^{-1}) for *c*. 8 or 1 hours, respectively; this procedure induced minimal aggregate slaking (MAS) or severe aggregate slaking (SAS), respectively. The saturated soils were then placed at a slope of 15% and immediately subjected to a 60 mm deionised water rainstorm, with an intensity of 36 mm hour^{-1}, a rainfall kinetic energy of 15.9 kJ m^{-3} and average droplet diameter of 2.97 ± 0.05 mm, using a drip-type laboratory rainfall simulator (Shainberg *et al*., 2003). Runoff samples containing sediments were collected continuously for each 6 mm rain interval of the storm in order to study the association between seal development and changes in the amount and PSD of the eroded material. Runoff volume and the mass of the dried eroded sediments were measured. The eroded sediments were then dispersed and analysed for their PSD. All treatments had three replicates.

Particle size distribution determination

We decided to disperse the eroded sediments and to study the primary PSD rather than examining the effective PSD of the non-dispersed sediment samples. This was because there was some concern, supported by experimental observations, that further breakdown of aggregated material could be induced during the procedure for PSD determination of the non-dispersed samples, which may have led to erroneous results and conclusions.

We also decided to use the laser diffraction technique for PSD determination because the laser diffraction method provides a continuous PSD rather than an arbitrary division of the particles among a limited number of size fractions (as determined by traditional methods based on sedimentation and/or sieving) and enables a more detailed data analysis of a desired size range, especially within the clay size fraction (Eshel *et al*., 2004). Furthermore, results from sedimentation-based methods (e.g. pipette or hydrometer) for particles <1 μm are increasingly unreliable because of the effect of Brownian motion on the rate of sedimentation (Allen, 1981; Loveland & Whalley, 2001). The laser technique makes the same assumption as standard methods that particles are spherical.

A Horiba LA-910 laser particle size analyser with a 632.8 nm He-Ne laser beam was used for determination of the detailed PSD of the soil primary particles, which passed through a 250 μm sieve. The instrument measures particle size over the range of 0.02–1000 μm.

The calculation module of the LA-910 (software version 1.31) uses the Mie theory. The Mie theory model requires, as an input parameter, the refractive index, which is a complex number comprising: (i) a real part (n_r), which represents the change in the velocity of light through the tested material compared with the velocity of light in a vacuum, and (ii) an imaginary term (n_i), which represents the transparency and absorptivity of that material. Following the study of Eshel *et al.* (2004) on the impact of the refractive index on the results for soil material, we chose to use $n_r = 1.52$ and $n_i = 0.2$ for the optical model calculations. The computed PSD data were expressed as volume percentages rather than mass percentages, as would be determined by sieving or sedimentation methods. Note that, for reasons given by Eshel *et al.* (2004), these values are not the same as those that would be determined by the hydrometer method (see Table 1, Eshel *et al*., 2004).

For each PSD analysis, 0.1–0.5 g of oven dried soil or sediment was shaken overnight in a 25 ml scintillation vial containing 10 ml of a 50 g l^{-1} sodium hexametaphosphate solution. The suspension was then transferred to a 250 ml beaker, and deionised water added to bring the volume to 100 ml. The 100 ml suspension was subjected to 60 s of ultrasonication and then passed through a 250 μm sieve. The material retained on the sieve was oven-dried and weighed.

The remaining suspended sediment (< 250 μm fraction) was collected in a 250 ml beaker, its volume made up to 200 ml with

Table 2 Measured total soil and clay losses during a 60 mm rain storm, and the enrichment of clay in the eroded material over the amount calculated to be lost when based on the assumption that it had the same percentage clay content as the parent soil, for severe aggregate slaking (SAS) and minimal aggregate slaking (MAS) treatments in the three soils studied

	Total soil loss		Total clay loss		Clay enrichment	
	SAS	MAS	SAS	MAS	SAS	MAS
Soil type	kg ha^{-1}					
Loam	10 253 ± 1123[a]	968 ± 18	1844 ± 202	963 ± 4	409 ± 47	203 ± 7
Sandy clay	10 838 ± 878	2115 ± 351	4843 ± 392	1044 ± 173	1018 ± 86	297 ± 45
Clay	4748 ± 277	168 ± 52	2367 ± 149	69 ± 24	440 ± 23	1 ± 0.3

[a]Mean value ± 1 standard deviation.

deionised water, and thoroughly mixed at high speed with a magnetic stirrer for 3 minutes. A sub-sample of 2–12 ml was then extracted in 2 ml aliquots from the stirred suspension and added to the fluid module of the laser instrument, which contained 240 ml of a base solution (0.4 g l^{-1} sodium hexametaphosphate and 0.01 g l^{-1} surfactant (Tween 21)). The variation in the amount of sub-sample transferred to the fluid module depended on the need to satisfy the light transmittance requirements of the laser analyser. The suspension in the fluid module was then subjected to a further 3 minutes of ultrasonication, to ensure complete dispersion of micro-aggregates, and 3 minutes of thorough mixing prior to three consecutive PSD measurements by the LA-910, taking 10–100 s each depending on the soil type.

Results and discussion

Runoff and soil loss

Similar to the results reported by Shainberg *et al.* (2003), greater infiltration rates were observed throughout the storm for samples of each soil type when MAS occurred than for those with SAS (i.e for samples that were saturated at the slow or fast wetting rate, respectively) (Figure 1). These greater infiltration rates indicate that seal development was less severe when surface aggregates underwent MAS rather than SAS. This effect was larger as soil clay content increased (Figure 1) because of associated greater aggregate stability (Levy & Mamedov, 2002). Consequently, all the soil samples with MAS generated less runoff than the same soil sample type with SAS. Correspondingly, total soil losses during the 60 mm rainstorm (Table 2) were significantly smaller in the case of MAS compared with SAS for all soils. The loam and sandy clay soils with SAS had similar sediment loads that were significantly greater than those observed from the clay soil (Table 2). For MAS-treated soils, sediment load was extremely small for the clay soil. This observation was expected, given the negligible amount of runoff and the large infiltration rate of this clay soil (Figure 1). It should be noted that, under our experimental conditions, runoff samples collected from all the soils contained a certain, albeit small, amount of material that was splashed directly into the runoff collectors rather than being removed by the runoff water alone. For the clay soil with MAS, this splashed material probably constituted the majority of the eroded material collected.

Total soil losses are directly attributable both to the total amount of runoff generated and to the rate of overland flow that directly affects the shear force and the carrying capacity of the runoff. However, even if total runoff volume had been equal for all the treatments, the differences in soil losses occurring from a given soil, subjected to either SAS or MAS, could not be explained entirely by differences in the runoff rates. Presenting sediment load as a function of runoff rate (Figure 2) demonstrates that lower sediment loads were removed from the soil subjected to MAS when compared with the loads from the same soil in the SAS case even when the runoff rate was of comparable magnitude. This suggests that the soil surfaces of MAS-treated soils were more cohesive and able to resist detachment than with SAS and/or that the amount of loose soil material that can readily be transported was more limited in the MAS soils than in the SAS ones.

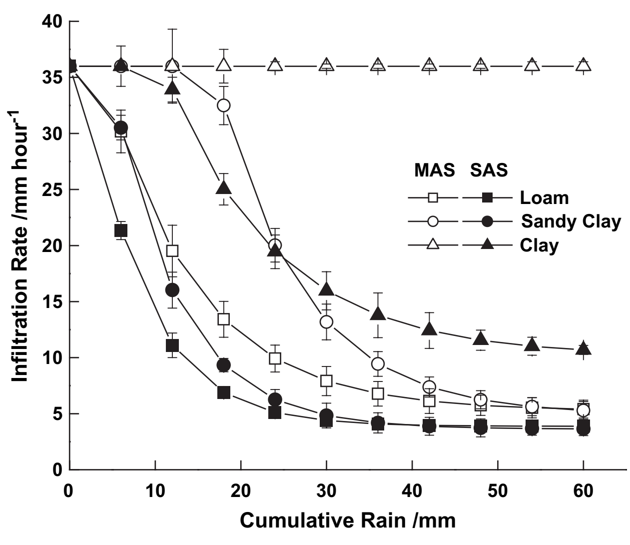

Figure 1 Effects of minimal aggregate slaking (MAS) and severe aggregate slaking (SAS) on infiltration rate.

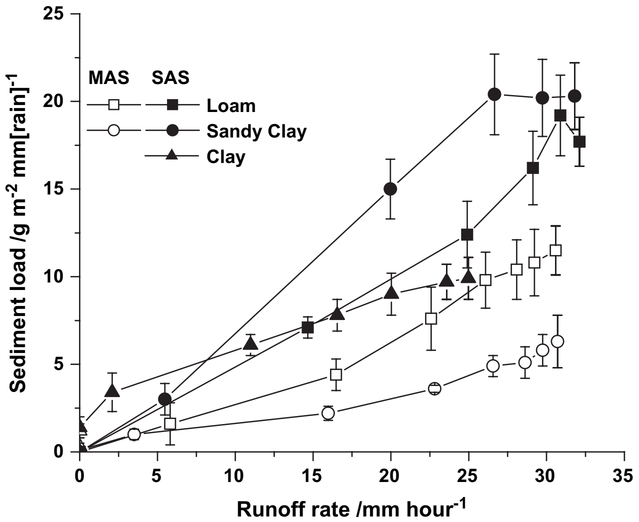

Figure 2 Effects of minimal aggregate slaking (MAS) and severe aggregate slaking (SAS) on sediment load as a function of runoff rate. Error bars represent ± 1 standard deviation. Note: for purposes of clarity, data points with very similar values within a given soil type and treatment are not shown.

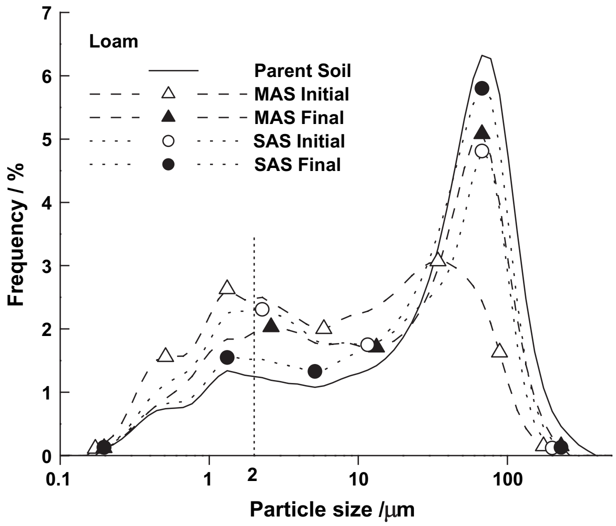

Figure 3 Laser diffraction derived size frequency distribution (presented as volume percentage) of particles <250 μm for the loam parent soil and the dispersed eroded material collected in the first and last 6 mm interval of rain from the samples subjected to minimal aggregate slaking (MAS) and severe aggregate slaking (SAS).

Particle size distribution (PSD) analyses

In the dispersed parent soil material, particles > 250 μm (medium and coarse sand) amounted to 2.3, 7.8 and 1.6% of the loam, sandy clay and clay soils, respectively. However, in the case of the eroded material, only negligible amounts (< 0.5%) of particles >250 μm were detected. The following discussion will, therefore, concentrate on the PSD of particles < 250 μm.

The primary PSD data of the <250 μm particles are presented as frequency curves for each soil separately in Figures 3–5. The figures compare the parent soil with the eroded material collected at the initial stage of seal development and when the seal was more developed (i.e. for the first and the final 6 mm rain intervals of the storm, respectively), for both degrees of aggregate slaking. The data clearly show that: (i) the three soils exhibited PSD frequency curves with different characteristic shapes, and (ii) within a soil type, the curves for both the eroded material and for the parent soil were, in general, of the same shape.

I. Loam. The PSD curves of the parent soil and of the eroded material were generally characterized by two main peaks, a minor peak, which occurred in the region of the coarse clay size fraction (1–2 μm), and a major peak, which occurred in the region of the fine sand size fraction, with a maximum frequency value for a particle size of *c.* 63 μm (Figure 3). It was further noted that, in the region of the minor peak: (i) the frequency curves for the eroded materials were greater (i.e. indicating larger fine material contents) than that of the parent soil, and (ii) the frequency curves for the eroded sediments during the initial stage of seal development were greater than those for the eroded sediments at the stage where the seal was fully developed. Opposite trends were observed in the region of the larger peak, indicating larger contents of coarse material in the parent soil compared with that in the eroded material. In addition, more coarse material was observed at the final stage of the storm compared with the initial one, and for SAS

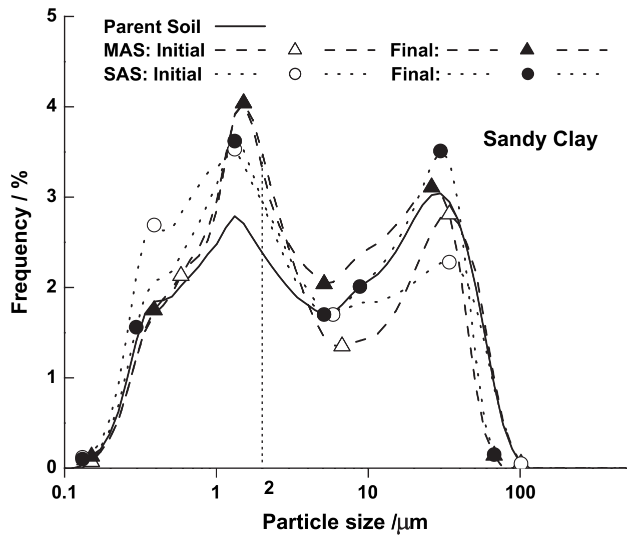

Figure 4 Laser diffraction derived size frequency distribution (presented as volume percentage) of particles <250 μm for the sandy clay parent soil and the dispersed eroded material collected in the first and last 6 mm interval of rain from the samples subjected to minimal aggregate slaking (MAS) and severe aggregate slaking (SAS).

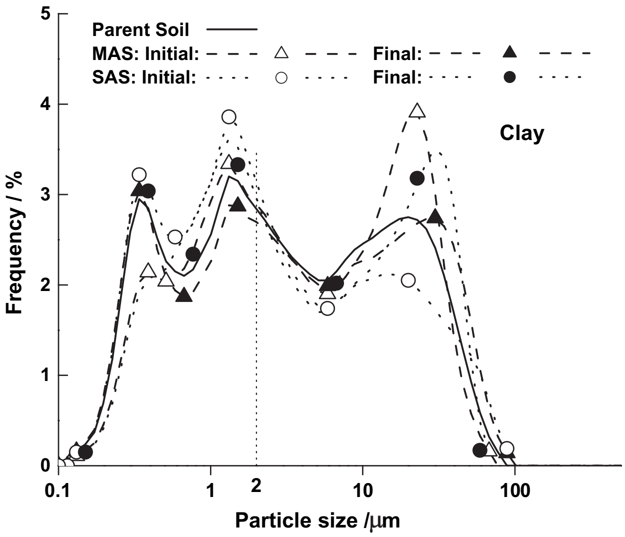

Figure 5 Laser diffraction derived size frequency distribution (presented as volume percentage) of particles <250 μm for the clay parent soil and the dispersed eroded material collected in the first and last 6 mm interval of rain from the samples subjected to minimal aggregate slaking (MAS) and severe aggregate slaking (SAS).

compared with MAS at any given stage of seal development (Figure 3).

II. Sandy clay. The PSD curves for the parent material and the eroded material were, similarly to the loam, characterized by two main peaks but of a comparable magnitude; one peak was in the region of the coarse clay size fraction (1–2 μm) and the other in the region of the silt to fine sand size fraction (20–50 μm) (Figure 4). A smaller subsidiary peak was also noted on the main clay peak in the fine clay fraction (~ 0.4 μm); this was most noticeable in the SAS curve. Similarly to the loam, there was more fine material in the eroded sediment than in the parent soil (i.e. the peaks in the region of the coarse clay fraction had the smallest magnitude for the parent soil) (Figure 4). Furthermore, the frequency curves for the MAS treatment (irrespective of rain depth) were larger in the region of this peak than those for the SAS treatment (Figure 4). In contrast, the peak in the region of the silt to fine sand size fraction followed a different trend, whereby the peak of the parent soil was below those of the eroded sediments collected during the final stage of the rain storm where seal development had reached an equilibrium, irrespective of the degree of aggregate slaking, but greater than those of the eroded material collected initially, when seal development was just commencing (Figure 4).

III. Clay. In contrast to those of the other two soils, the PSD curves for the parent soil and the eroded material were characterized by three peaks of a similar size (Figure 5); maximum frequencies occurred in the region of the fine clay (0.3 μm), coarse clay (1.3 μm) and silt (20 μm) size fractions. For all three peaks, the frequency curve of the parent soil occupied an intermediate position between the curves of the eroded material (Figure 5). In contrast to the other two soils, no noticeable trends could be observed with respect to the impact of the degree of aggregate slaking and/or the degree of seal development on the frequency curves.

Changes in clay content affected by degree of aggregate slaking

In an attempt to make a more detailed and quantitative evaluation of the effects of the degree of aggregate slaking and of the degree of seal development on the PSD of the eroded sediments, we examined the differences in the clay-sized fraction (<2 μm) more closely. We first looked at the percentage change in clay content in the eroded material relative to its content in the parent soil. This quantity, termed the relative change in clay content (Δc), was computed for each 6 mm rain interval for all of the treatments studied as follows:

$$\Delta c = 100(Ec - Pc)/Pc, \tag{1}$$

where Ec and Pc are clay contents of the eroded material and parent soil, respectively, expressed as % by volume. Thus, positive values for Δc indicate that the eroded material was enriched by clay material relative to the parent soil whilst negative values indicate that the eroded material was depleted of clay material relative to the parent soil. In addition, we determined the excess amount of clay lost over the entire 60 mm rainstorm by subtracting the mass of clay that would have been present in the eroded material, had it had the same textural composition as the parent soil, from the actual mass of clay determined to be in the material. These totals were referred to as the cumulative enrichment of clay in the eroded material (CUMc) and were expressed in kg ha^{-1}.

The Δc of the eroded material differed among the three soils and depended on the degree of aggregate slaking. In the SAS treatments, positive values were obtained for Δc for all three soils (i.e. there was enrichment in clay material in the eroded sediments relative to the parent soil) (Figures 6–8). The CUMc for the entire 60 mm of rain in the sandy clay was more than twice that observed in the loam and clay soils for the SAS treatment (Table 2). These levels of cumulative clay enrichment represent, in the case of the SAS treatment, an increase in clay loss of 28.5, 26.6 and 22.8% for the loam, sandy clay and clay, respectively, over the amount of eroded clay that would have been obtained had the eroded material possessed the same clay content as the parent soil (Table 2). These substantial percentages of increased clay losses suggest that some soil degradation would occur with respect to nutrient losses, reduced aggregate strength and, subsequently, poorer soil structure at the point of origin of the eroded material. Furthermore, these percentages of cumulative clay enrichment in the eroded material were inversely related to the parent soil

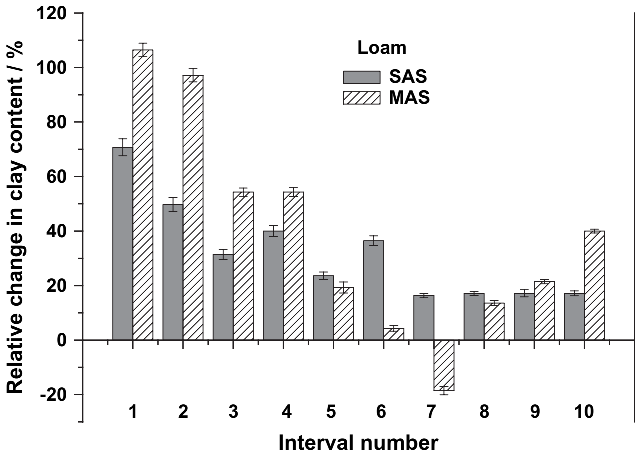

Figure 6 Relative change in clay content in the eroded material from the loam soil with minimal aggregate slaking (MAS) and severe aggregate slaking (SAS) occurring over 10, 6 mm rain depth intervals. Error bars represents ± 1 standard deviation.

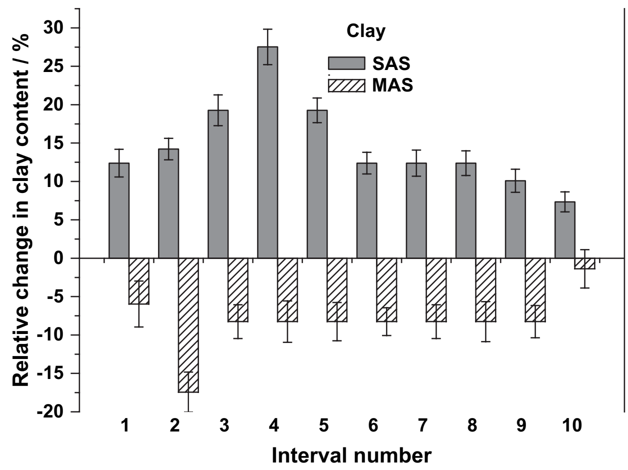

Figure 8 Relative change in clay content in the eroded material from the clay soil with minimal aggregate slaking (MAS) and severe aggregate slaking (SAS) occurring over 10, 6 mm rain depth intervals. Error bars represent ± 1 standard deviation.

clay content, 14.0, 35.4 and 43.6% (determined by the traditional hydrometer method) for the loam, sandy clay and clay, respectively (Table 1). Soil clay content is known to affect aggregate stability, with the latter commonly decreasing with the decrease in clay content (Kemper & Koch, 1966; Levy & Mamedov, 2002). This suggests that the susceptibility to produce eroded material enriched in clay could be directly related to the aggregate stability of the soil. Aggregates with a greater tendency to slake by fast wetting and/or break down by the impact of raindrops, produce aggregate fragments that, with their greater specific surface areas, can more readily release clay size particles into the runoff water, thus leading to greater

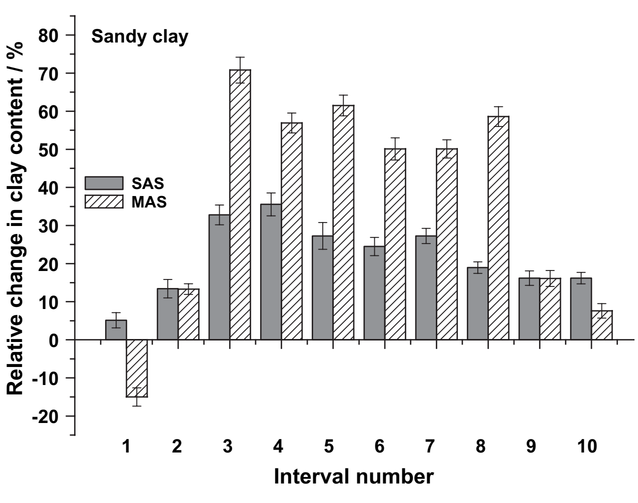

Figure 7 Relative change in clay content in the eroded material from the sandy clay soil with minimal aggregate slaking (MAS) and severe aggregate slaking (SAS) occurring over 10, 6 mm rain depth intervals. Error bars represent ± 1 standard deviation.

clay enrichment than in the case of more stable aggregates. Furthermore, a soil already comprising unstable aggregates would be more susceptible to aggregate, and hence structural, degradation by this process.

Generally, in both the MAS and the SAS treatments, positive Δc values were obtained in the loam and the sandy clay soils (Figures 6, 7). In addition, the Δc values for the MAS treatment were usually similar to or greater than those of the SAS treatment. The CUMc at the end of the storm in the MAS treatment for the sandy clay was somewhat greater than that for the loam (Table 2). However, these CUMc values were considerably lower than the corresponding values for the SAS treatment in these two soils, and confirmed our initial expectation that the SAS treatment would result in greater losses, not only of the total soil (Table 2) but also of clay size particles, than the MAS treatment. Similar to observations made by Levy *et al.* (1997), SAS, because of fast wetting, results in larger numbers of smaller broken aggregates at the soil surface, faster development of a more impermeable seal (Figure 1) and thus a greater load of eroded material in the runoff (Figure 2; Table 2).

Our results for the loam and sandy clay indicate comparable, and frequently larger, Δc values in the MAS than in the SAS treatment, but larger CUMc values in the SAS compared with the MAS treatments (Figures 6, 7; Table 2). This apparent discrepancy can be explained as follows. The SAS treatment results in larger amounts of runoff, which have a higher carrying capacity capable of transporting larger amounts of soil (Figure 1). The sediment in the runoff from the SAS-treated soils thus contains greater amounts of all size fractions, including the clay size particles, resulting in larger CUMc values for this treatment compared with those of the MAS treatments. However, the higher carrying capacity also results in a greater proportion of coarser material carried both as primary particles and as small aggregates. The increase in the proportion of coarse primary particles leads to lower values of Δc for the SAS treatment when compared with MAS. Thus, enrichment with clay occurs at a lower rate from the SAS than from the MAS treatment but the amount of excess clay transported is substantially increased.

The effects of MAS on the PSD of the eroded material from the clay differed from its effects on the eroded material from the other two soils. In contrast to the loam and sandy clay, MAS in the clay yielded negative Δc values throughout the storm, indicating depletion of clay particles in the eroded material relative to the parent soil; the opposite was true for the SAS treatment (Figure 8). Furthermore, unlike the situation in the other two soils, MAS had a negligible impact on the CUMc in the eroded material (<-2 kg ha^{-1}), representing <3% decreased loss of clay than would have been the case had the eroded material possessed the same clay content as the parent soil (Table 2). These differences in the response of the clay when subjected to MAS, compared with the other two soils, could be ascribed to the means by which the eroded material was transported. Whereas in all the other treatments the eroded material had been transported mainly by runoff, in the case of the clay subjected to MAS, where the infiltration rate was comparable to the rain intensity (Figure 1) and, therefore, very little runoff was generated, the eroded material was mainly splashed directly into the runoff collectors rather than being removed by the runoff water.

The degree of seal development also affected the PSD of the eroded material. In general, the percentage content of the clay and coarse fractions of the sediments were closer to those of their respective parent soils in the final stage of the storm (Figures 3–5), where seal development had generally reached equilibrium, than in the initial stage, where seal development was commencing, which was in agreement with the observations of Mitchell *et al.* (1983).

For the loam, most of the enrichment of the eroded material by clay took place during the first 24 mm of rain (i.e. the first four intervals of 6 mm of rain, Figure 6), which corresponded with the depth of rain needed for: (i) most of the seal development to occur (Figure 1), and (ii) the attainment of a constant load of sediments in the runoff water (Figure 2). Once the seal approached full development (based on its asymptotic approach to a steady state infiltration rate, Figure 1) the difference between the clay content in the eroded material and that of the parent soil material was much smaller (Figure 6). These observations support the conclusion that it was the presence of easily broken aggregates in the loam that was the main driving force for the enrichment of claysize particles in the eroded sediments. With increased depth of rain and the formation of the seal, fewer of these aggregates were available for breakdown as loose material and became incorporated into the seal. Furthermore, the structure of a seal, as described by McIntyre (1958), results in a compacted upper layer comprising coarse material, which would reduce the availability and thus the preferential removal of clay particles. This is in agreement with the data presented in Figure 6, showing that the enrichment of the eroded material by clay particles is reduced after the seal approaches full development (i.e. during intervals 5–10 (30–60 mm of rain)).

In the sandy clay, much of the clay enrichment of the eroded material took place during intervals 3–8 (18–48 mm of rain) (Figure 7) with maximum clay enrichment occurring in the third and fourth intervals for the MAS and SAS treatments, respectively. In contrast, the maximum clay enrichment in the loam took place in the first interval for both treatments (Figure 6). In the MAS treatment of the sandy clay, the clay enrichment of the eroded material took place, as with the loam, during the rain intervals when the seal was being developed (Figure 1). Clay enrichment sharply decreased after the seal approached full development in the ninth interval (Figure 1). Conversely, in the SAS treatment, the enrichment of eroded material with clay increased to a maximum in the fourth interval, which corresponded with the seal approaching full development (Figure 1). Thereafter, when sediment load in the runoff water became more or less constant (Figure 2), clay enrichment decreased steadily to approximately the same level exhibited by the MAS treatment in the final stages of the rainstorm (Figure 7).

In the clay subjected to SAS, the relative change in clay content in the eroded material increased until the fourth rain interval and thereafter decreased (Figure 8). This behaviour could neither be linked to specific stages in seal development (Figure 1), nor to changes in sediment load (Figure 2). In the MAS treatment, the relative change in clay content was constant throughout most of the rain storm (Figure 8) and could be explained, as mentioned previously, by the lack of development of a seal, resulting in large infiltration rates, (Figure 1), correspondingly small runoff rates, and, therefore, the small amounts of sediments (Table 2) were mainly the result of transportation by splash.

As noted above, the pattern of relative clay enrichment in the eroded sediments from the sandy clay and clay soils differed markedly from that in the loam. These differences could be explained by the nature of the seals developed by the soils after SAS. In the presence of a fully developed seal, the nature of the soil material that can be detached and transported will be determined by the particles comprising the upper layer of the surface seal and by how readily these particles can be removed. The upper layer of the surface seal is composed of a compact layer of coarse micro-aggregates and primary particles (McIntyre, 1958). Additionally, in the final stages of seal development, there exists a balance between seal formation and seal destruction processes, with the latter, attributed to erosion, exposing subsurface soil particles (Poesen, 1986). In the case of the loam, the weak surface aggregates are rapidly broken down into microaggregates, which are expected to have similar PSDs to that of the parent soil and thus do not contribute to the phenomenon of clay enrichment, and also into primary particles. The small clay content of the soil means that relatively small amounts of eroded clay-size particles result in initially large values of Δc that diminish rapidly over time as the clay size particles are washed from the soil surface by runoff leading to lower rates of preferential clay erosion (Figure 6) even in the presence of a partially developed seal (Figure 1), including one case where Δc became negative. Seal destruction processes ensure that clay size particles do, however, continue to be available throughout the storm.

In the finer-textured sandy clay and clay soils, the larger fractions of clay size particles in the parent material led to lower initial values of Δc than those for the loam, and these diminished less rapidly with time because greater amounts of clay size particles were available to be removed in the runoff. Therefore, preferential removal of clay was not immediately reduced (Figures 7, 8) by the formation of the seal (Figure 1). Furthermore, seal destruction processes would provide greater quantities of clay-size particles than in the case of the loam. It is possible that the proportion of coarse primary particles to micro-aggregates that were susceptible to erosion, also changed in favour of the former, so that preferential clay erosion was gradually reduced over time (Figures 7, 8). Note that during the first interval of the MAS treatment, when the seal was relatively undeveloped compared with that of the SAS treatment, coarse size rather than clay size primary particles were preferentially removed (Figure 7). This can be ascribed to the clay-size particles resulting from dispersion at that stage being preferentially washed into the underlying soil pores rather than being removed in the runoff water.

Two additional means by which relative clay enrichment may be reduced during a rainstorm are postulated: (i) reduction of clay dispersion from aggregates at the soil surface resulting from the absorption of raindrop impact by increased runoff water depth (Ferreira & Singer, 1985), and (ii) increased availability of more readily detached coarser material, including fine sand particles (50–250 μm), on the exposed mounds that form between the shallow channels during the rainstorm (Levy *et al*., 1988). These latter features may also contribute to the initially larger levels of clay enrichment and hence coarse material depletion because, during their formation, they may represent coarser material that is removed from the runoff stream. Furthermore, after their formation they shield the underlying soil, reducing further aggregate breakdown and thus the release of available clay-size particles (Hairsine *et al*., 1999).

Conclusions

Characterization of the PSD of eroded sediments from three smectitic soils varying in texture showed that, in general, there was an enrichment of clay material in the eroded sediments relative to the parent soil. This observation was in accordance with the findings of Monke *et al*. (1977) and Alberts *et al*. (1983). In the case of the SAS treatment, the cumulative enrichment in clay material was inversely related to soil clay content, which suggested that this enrichment could be linked to aggregate stability. MAS in the loam and sandy clay resulted in greater relative enrichment of clay material in the eroded sediment compared with SAS, even though the latter treatment caused and enhanced aggregate slaking and disintegration throughout the rainstorm. The SAS treatment resulted, however, in much greater levels of cumulative clay enrichment than the MAS treatment because of greater amounts of total soil erosion. With respect to the effects of depth of rain (i.e. degree of seal development) on the PSD of the eroded sediments, in some cases the enrichment of the eroded material by clay-size particles took place mostly during the stage of rain at which the seal was beginning to develop (i.e. loam and sandy clay subjected to MAS); in others (clay and sandy clay subjected to SAS), the accumulation of clay-size material in the eroded sediments continued throughout the course of seal development.

The observed enrichment of the eroded material by clay-size particles and its dependence on the degree of aggregate slaking (SAS vs. MAS) amplifies the importance of protecting surface aggregates from breaking down during rainstorms. It also re-emphasizes, especially in the SAS treatment, the hazard of eroded sediments as a potential source of pollution, degrading water quality in river systems and contaminating downstream areas. A possible management technique to reduce soil loss from agricultural land may involve irrigating a soil in short pulses to induce MAS prior to more intensive irrigation or a forecast rainstorm.

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